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Hudson, J. L.

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University of Virginia
Office of Sponsored Programs
P. O. Box 400195
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14. ABSTRACT

We carried out experiments & supporting simulations on the dynamics of nonlinear complex systems with coupling, feedback, external forcing, and noise. The experiments are carried out with arrays of electrodes where: (i) The sites have measurable rates of reaction & are individually addressable; (ii) each site has rich dynamics depending on reactor parameters; (iii) the strength & length of scales of coupling among the elements are controllable.

We have provided a proof of the main predictions of theory on the emergence of synchronization of coupled smooth oscillators; we demonstrate a phase transition & the dependence of order on coupling strength predicted by the theory. Furthermore, we have shown a strong enhancement of fluctuations near the critical point & have shown that the principal predictions hold also for relaxation & even weakly chaotic oscillators that often occur in physical systems. For very weak coupling of chaotic systems there are no qualitative effects on the local chaotic dynamics. We have shown that these small effects can nevertheless produce significant changes in the collective, or overall, behavior of the system & that the collective behavior can be quite different from the local behavior.

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Supporting simulations, array of electrodes, synchronization of coupled smooth oscillators & chaotic oscillators

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Final Report

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Principal Investigator	John L. Hudson
Organization	University of Virginia

Program Officer: Dr. Michael Shlesinger
May, 2003.

Technical Section

Technical Objectives

Our goal was to explore the dynamics of nonlinear complex systems with coupling, feedback, external forcing, and noise. Engineering of chemical complexity can lead to optimal local and global chemical reaction rates in production and in power generation. Special attention was dedicated to synchronization properties of laboratory systems with inherent heterogeneities. Synchronization of oscillators may increase power of lasers, improve communication, and aid in understanding biological systems.

Technical Approach

A chemical reactor consisting of an array of electrodes has been designed with the following characteristics: (i) the sites have measurable rates of reaction and are individually addressable; (ii) each site has rich dynamics (periodic, chaotic) depending on reactor parameters; (iii) the strength and length scales of coupling among the elements are controllable; (iv) feedback control and external forcing or noise can be applied; (v) heterogeneities among elements model the characteristics of engineering and natural systems.

Progress

During the grant period we made progress in several areas with support from ONR.

Emerging Coherence in a Population of Chemical Oscillators

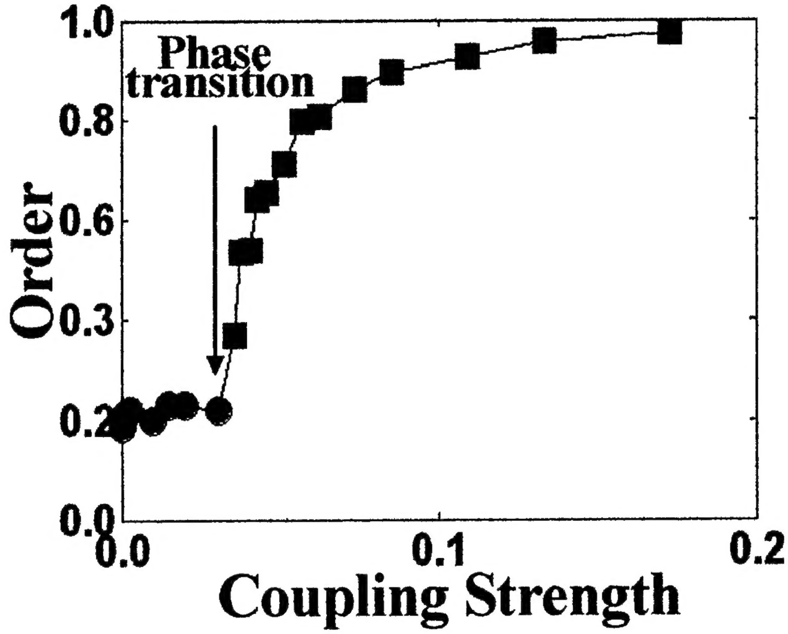
The theory of synchronization of coupled oscillators developed in the 1970s by Winfree and Kuramoto has played a fundamental role in the development of a field of nonlinear science dealing with collective dynamics. Applications in engineering may be found not only in coupled chemical reactions, but also in other areas such as microwave systems, lasers, and digital logic circuitry. It has also been

shown to be an important process in the functioning of heart pacemaker cells and of yeast cells as well as in the synchrony of flashing fireflies and chirping crickets. However, a systematic experimental verification has been missing. Using as an example a population of globally coupled electrochemical oscillators, we have provided a proof of the main predictions of the theory; we confirm the phase transition and the dependence of order on coupling strength predicted by the theory (Fig. 1). Furthermore, we have shown a strong enhancement of fluctuations near the critical point and have shown that the principal predictions hold also for relaxation and even weakly chaotic oscillators that often occur in physical systems. [3]

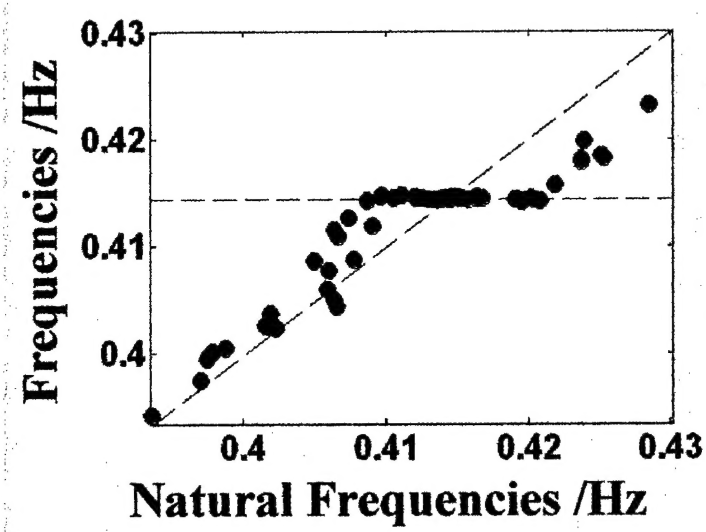
Collective dynamics of weakly coupled chaotic electrochemical oscillators
In chemically reacting systems the rate of reaction is often a function of both space and time. The spatial scale of variations in fluid/solid or electrochemical systems can range from nano- through micro- to macroscopic and variations at several scales can occur simultaneously, e.g., small-scale on a catalyst surface up to the larger scale of the reactor. The degree of interaction among the reacting sites is influenced both by the local reaction rate and the strength and range of the coupling. We have carried out experimental studies on electrochemical reaction sites in which the coupling is weak so that there are no qualitative effects on the local chaotic dynamics. We have shown that these small effects can nevertheless produce significant changes in the collective, or overall, behavior of the system and that the collective behavior can be quite different from the local behavior; for example, even when the local dynamics are chaotic the collective behavior can be periodic or almost constant (Fig. 2). Phase synchronization obtained at weak coupling strengths is shown in Fig. 3; for comparison, behavior with stronger coupling is also shown. In chemically reacting systems this spatially averaged reaction rate is usually the quantity of interest since it determines the overall conversion in the reactor. [4, 8]

Organizing spatiotemporal patterns with global interactions: coupling, forcing, and feedback
Experiments were carried out on arrays of chaotic electrochemical oscillators to which global coupling, periodic forcing, and feedback were applied. The characteristics of the three methods in the development of structures are compared. Global coupling, as well as forcing and feedback control applied to weakly coupled systems, synchronizes populations of chaotic oscillators. More complicated structures involving the existence of clusters of states have also been found in the experiments; many stable structures can arise under a given set of conditions. States with from two to four co-existing cluster states have been found. By controlling the external variations or system feedback desired states can be obtained (Fig. 4). Such studies on the engineering of chemical complexity, that is, using coupling, feedback control, external forcing, and added noise to influence spatiotemporal patterns and system behavior, can lead to optimized behavior of chemical processes. Furthermore, an external forcing signal on nonlinear oscillators can simulate periodic environmental variations on complex systems such as those found in biological contexts. [2,10]

Fig. 1. Emerging coherence in a population of nonidentical periodic oscillators.



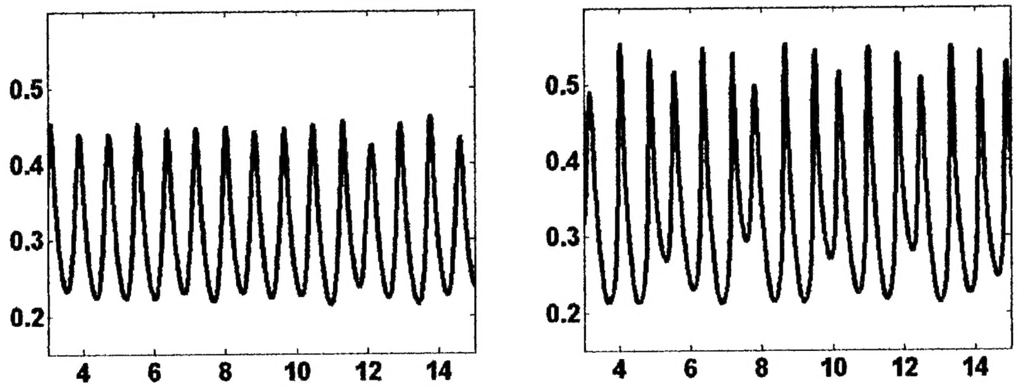
a. With increasing the global coupling strength among the oscillators a phase transition occurs above which order emerges.



b. At coupling strength just above the phase transition (shown here) the oscillators with frequencies close to the mean frequency are locked.

Fig. 2. Collective dynamics of population of chaotic oscillators

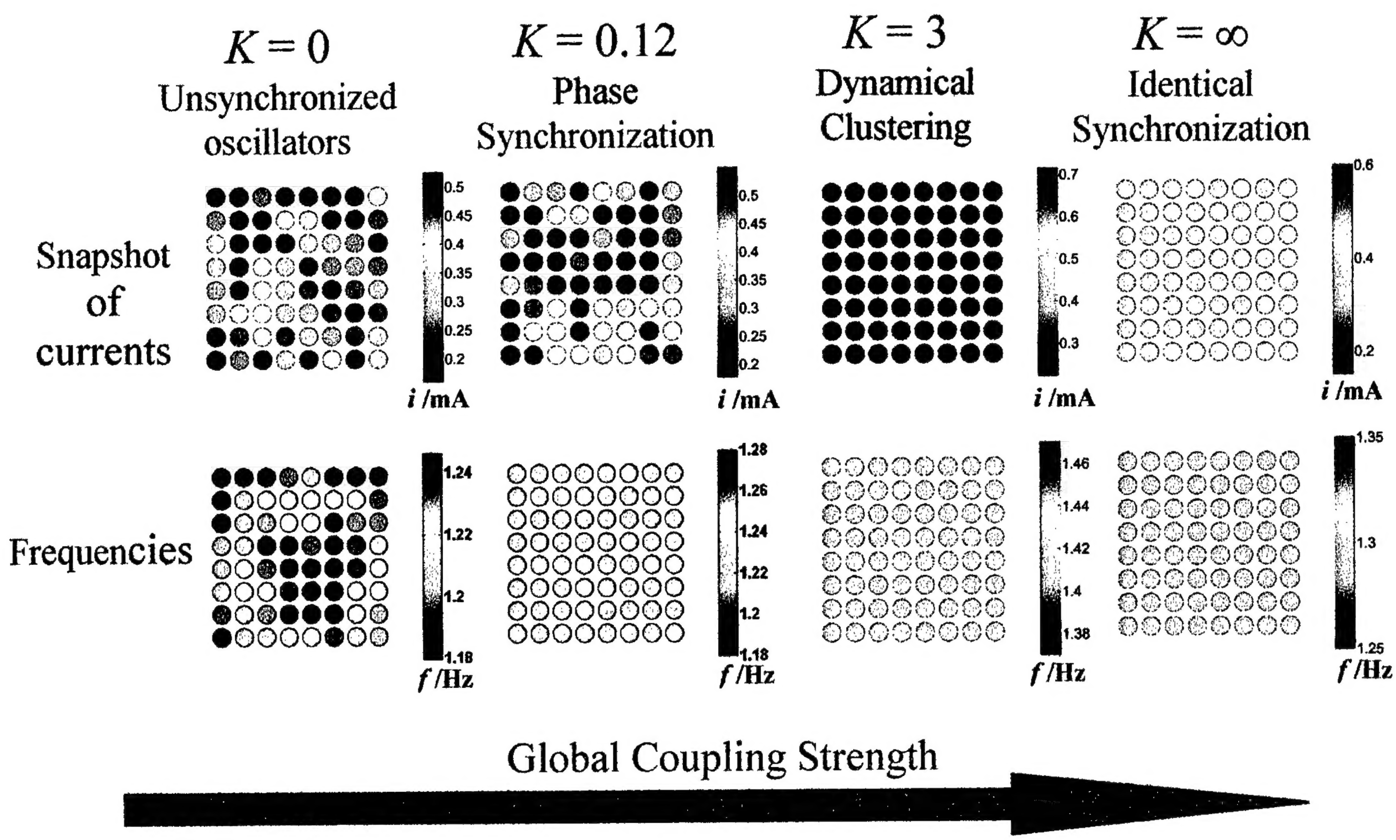
The collective (spatially averaged) behavior of populations can be qualitatively different form the local behavior. In the example shown here, the global nearly periodic oscillations of averaged Ni dissolution rate are produced with chaotic variations of local rates.



a. Average current vs. time. b. Local current vs. time.

Fig. 3. The effect of global coupling on populations of chaotic oscillators

With increasing the global coupling strength (K) transitions are seen from unsynchronized through phase synchronized and clustered states to identical synchronization.



The effects of coupling and forcing on the synchronization characteristics of small sets of chaotic electrochemical oscillators

The effect of external forcing of a single chaotic chemical oscillator has been investigated: phase synchronization and suppression of chaos through intermittency have been observed depending on the amplitude and frequency of the forcing. Phase synchronization has been experimentally confirmed in a coupled two-oscillator system. Experimental tests of different forms of synchronization of such small sets of oscillators are useful since these have been proposed as ways of communication and encrypting. [1,5]

The constructive effects of noise on the synchronization characteristics of chaotic electrochemical oscillators

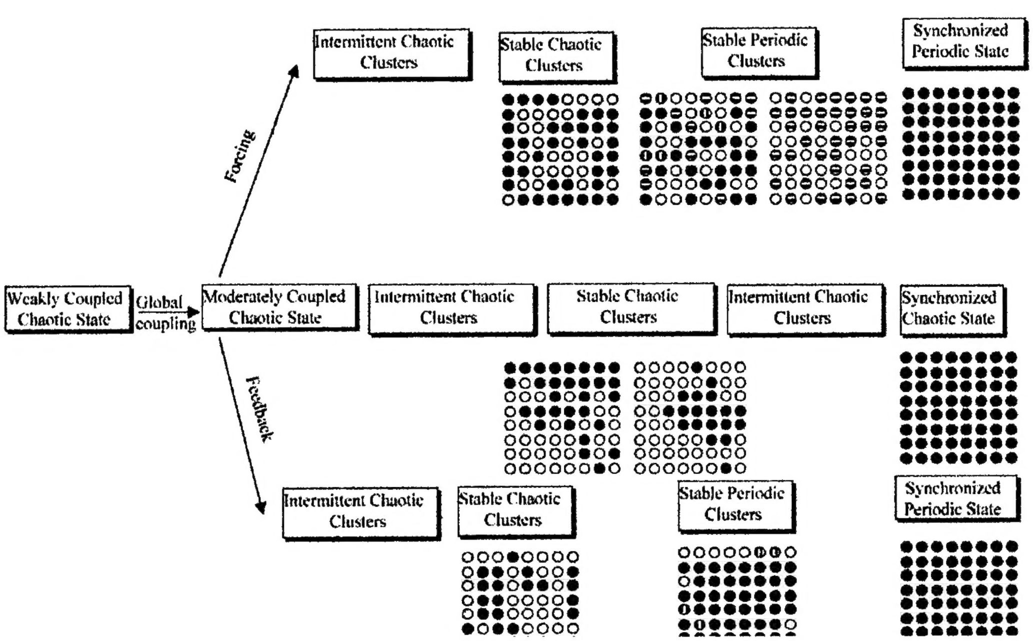
The effects of noise on synchronization have been studied in both small sets and in larger populations of coupled chaotic oscillators. In the region below phase synchronization noise has been found to enhance the extent of phase synchronization. The constructive effect of noise is interpreted by the synchronization properties of unstable periodic orbits embedded in the chaotic attractor. The exploration of the counterintuitive effects of noise on two coupled oscillators is important for experimental implementation of communication techniques. With larger populations the findings are significant in understanding the cooperative effects of noise and weak coupling in physical, chemical, and biological systems.; for example, environmental fluctuations may play a role in synchronization of population oscillations over large geographical regions. [6,9]

Stabilizing and tracking unknown steady states of dynamical systems

An adaptive dynamic state feedback controller for stabilizing and tracking unknown steady states of dynamical systems was proposed. We proved that the steady state can never be stabilized if the system and controller in sum have an odd number of real positive eigenvalues. For two dimensional systems, this topological limitation states that only an unstable focus or node can be stabilized with a stable controller and stabilization of a saddle requires the presence of an unstable degree of freedom in a feedback loop. The use of the controller to stabilize and track saddle points (as well as unstable foci) is demonstrated both numerically and experimentally with an electrochemical Ni dissolution system. [7]

Fig. 4. Engineering chemical complexity.

A wide variety of patterns are obtained with coupling, forcing or feedback.



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Kiss, István Z., Wen Wang, and John L. Hudson, "Forcing, Coupling, and Feedback of Chaotic Electrochemical Oscillators," Proceedings, 6th Experimental Chaos Conference, AIP Conference Proceedings *622*, 3-11 (2002).

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Institut für Physik, Humboldt-Universität zu Berlin, July 15, 2002.
Department of Chemical Engineering, University of Texas, October 22, 2002.

Presentations, Technical Meetings:

"Forcing, coupling, and feedback of chaotic electrochemical oscillators," 6th Experimental Chaos Conference, Potsdam, Germany, July 22-26, 2001.
"Phase Synchronization and Suppression of Chaos in Forcing of an Electrochemical Oscillator," poster, Gordon Research Conferences on Nonlinear Science, South Hadley, Massachusetts, Aug. 2001. Kiss, I. Z. and Hudson, J.L.
"Clustering of Arrays of Chaotic Chemical Oscillators by Feedback and Forcing", poster, Gordon Research Conferences on Nonlin1ear Science, South Hadley, Massachusetts, Aug. 2001. Kiss, I. Z., Wang, W., and Hudson, J.L.
"Synchronization of populations of chaotic electrochemical oscillators," International Workshop and Seminar on Control, Communication, and Synchronization in Chaotic Dynamical Systems, Max-Planck-Institute for Physics of Complex Systems, Dresden, Germany, Nov. 23, 2001.
"Emerging coherence in a population of chemical oscillators," Symposium on Engineering of Chemical Complexity, Fritz-Haber-Institut, Berlin, June 5, 2002.
"Collective Dynamics of a Population of Chemical Oscillators," Workshop on Pattern and Waves: theory and applications," St. Petersburg, Russia, July 8, 2002.
" Phase synchronization and collective dynamics of populations of chaotic electrochemical oscillators." poster, 7th Experimental Chaos Conference, San Diego, August 26-29, 2002. Kiss, I.Z., Y. Zhai, and J.L. Hudson
"Emerging Coherence and Collective Dynamics of a Population of Chemical Oscillators," Synchro 2002, Saratov, Russia, Sept. 23, 2002.
"Emerging Coherence and Collective Dynamics of a Weakly Coupled Electrochemical Reaction on an Array," AIChE 2002 Annual Meeting, Indianapolis, November 3-8, 2002. Istvan Z. Kiss, Yumei Zhai, and John L. Hudson.

Abstract

We carried out experiments and supporting simulations on the dynamics of nonlinear complex systems with coupling, feedback, external forcing, and noise. The experiments are carried out with arrays of electrodes where: (i) The sites have measurable rates of reaction and are individually addressable; (ii) each site has rich dynamics depending on reactor parameters; (iii) the strength and length scales of coupling among the elements are controllable.

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We also carried out experiments on the application of global coupling, periodic forcing, and feedback to arrays of chaotic electrochemical oscillators. By controlling the external variations or system feedback to influence spatiotemporal patterns and system behavior, desired states (synchronized behavior, cluster formation, etc.) can be obtained. The constructive effects of added noise on synchronization have been studied in both small sets and in larger populations of coupled chaotic oscillators.